

TITLE: INTEGRATED REMOVAL OF NO_x, WITH CARBON MONOXIDE AS REDUCTANT, AND CAPTURE OF MERCURY IN A LOW TEMPERATURE SELECTIVE CATALYTIC AND ADSORPTIVE REACTOR

AUTHORS: Neville G. Pinto (PI), Panagiotis G. Smirniotis (co-PI), and Stephen W. Thiel

STUDENT: Lei Ji

INSTITUTION: University of Cincinnati
Department of Chemical and Materials Engineering
University of Cincinnati
Cincinnati, OH 45221-0012

PHONE NO: (513) 556-4130

FAX NO: (513) 556-0128

EMAIL: Neville.Pinto@uc.edu

GRANT NO: DE-FG26-06NT42712

PERIOD OF PERFORMANCE: March 2007-February 2008

DATE: April 15, 2008

1. ABSTRACT

OBJECTIVES

The broad objective of this project is to develop a novel, advanced Low Temperature Selective Catalytic and Adsorptive Reactor (LTSCAR) for the simultaneous removal of NO_x and mercury (elemental and oxidized) from coal-fired flue gases in a single unit. It is anticipated that the proposed system will lower removal costs for the targeted pollutants by an order of magnitude relative to current technologies. Specific project tasks include development of a sulfur-tolerant supported metal oxide catalyst for low temperature SCR (LTSCR) using CO as a reductant; evaluation of LTSCR catalysts as mercury adsorbents; evaluation of the performance of nanostructured chelating adsorbents for mercury capture at LTSCR conditions; and evaluation of the performance of prototype LTSCAR systems.

ACCOMPLISHMENTS TO DATE

Catalyst Development. It was previously reported that 10 wt % MnO_x supported on titania is effective for LTSCR at industrially relevant conditions (50,000 hr⁻¹ space velocity, 2 vol % O₂). Unfortunately, this catalyst was not sulfur tolerant. In the current project year, a reaction mechanism has been developed for this catalyst, and key steps of the mechanism have been validated using FTIR. Sulfur tolerance has been addressed through development of catalysts consisting of MnO_x supported on mixed metal oxides (ceria+titania).

Supported Metal Oxide Adsorbents. It was previously reported that MnOx supported on titania captures elemental mercury at LTSCR temperatures, and preliminary work was completed on developing a mechanism describing Hg capture. MnOx/TiO₂ catalysts were not sulfur tolerant. In the current project year, additional mechanism development has been completed that suggests that the Hg adsorption rate is mass transfer limited. Preliminary tests using MnOx supported on CeO₂+TiO₂ indicate that these catalysts also capture Hg⁰, and are less susceptible to SO₂ inhibition.

Nanostructured Chelating Adsorbents. It was previously reported that nanostructured chelating adsorbents can capture Hg⁰ at LTSCR temperatures; the effectiveness of a number of ionic liquids using the bis(trifluoromethane sulfonyl) imide anion were reported. This work has been extended to the corresponding ionic liquids using chloride anions, and it was found that [bmim]Cl coated on MPTS-functionalized silica is effective for capture of both elemental and oxidized mercury at flue gas conditions. The development of an adsorbent with high capacity for both Hg(II) and Hg⁰ is a significant development milestone in control of mercury in flue gas.

FUTURE WORK

Future project efforts will be focused as follows:

- *Catalyst Development.* Engineer sulfur tolerance into the LTSCR catalyst; testing catalysts using simulated flue gas using CO as the reductant.
- *Supported Metal Oxide Adsorbents for Hg Capture.* Develop a mechanistic explanation of the support effect on sulfur tolerance; evaluate Hg capture using full simulated LTSCR gas compositions.
- *Nanostructured Chelating Adsorbents for Hg Capture.* Establishing the practical operating temperature range of the adsorbents; test combined Hg(II)+Hg⁰ capture with full simulated LTSCR gas compositions (pre- and post-LTSCR).
- *Combined Capture.* To date, the LTSCAR experiments have not been completed. During the next project year, a prototype LTSCAR system will be constructed and tested using industrially realistic conditions (composition, temperature, and space velocity).

2. JOURNAL ARTICLES, COMPLETED PRESENTATIONS AND STUDENTS RECEIVING SUPPORT FROM THE GRANT

Journal Articles

- L. Ji, S.W. Thiel, N.G. Pinto, "Room Temperature Ionic Liquids for Mercury Removal from Flue Gas," *Industrial & Engineering Chemistry Research*, submitted (2008).
- L. Ji, P.M. Sreekanth, P.G. Smirniotis, S.W. Thiel, N.G. Pinto, "Manganese Oxide/Titania Materials for Removal of NO_x and Elemental Mercury from Flue Gas," *Energy and Fuels*, accepted (2008).
- P. M. Sreekanth, P.G. Smirniotis, "Selective Reduction of NO with CO over Titania Supported Transition Metal Oxide Catalysts", *Catalysis Letters*, 22 (2008) 37-42.

Conference Presentations

- L. Ji, S.W. Thiel, P.G. Smirniotis, N.G. Pinto, "Room Temperature Ionic Liquids for Mercury Capture from Flue Gas," Poster presented at Department of Energy 2007 Mercury Control Technology Conference (December 2007).
- L. Ji, S.W. Thiel, N.G. Pinto, "Removal Of Mercury From Flue Gases Using Ionic Liquid Coated Chelating Adsorbents," AIChE 2007 National Meeting (November 2007).

Students Supported Under this Grant

- Lei Ji, Ph.D. student, Department of Chemical and Materials Engineering, University of Cincinnati. Degree to be granted June 2008; employed at Babcock and Wilcox May 2008.